Optimization of high-performance field emission rare earth tungsten alloy cathodes*

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The cathodes, as the electronic emission source of all kinds of electronic vacuum devices and spacecraft potential control system, its performance not only affects the overall efficiency of the equipment, but also limits the most important factors of the long life and high reliability of the system, and its emission principle mainly includes thermal emission and field emission, etc. Therefore, based on first-principles calculations using density functional theory, this study constructs atomic models of W cathode surfaces doped with different rare earth atoms. Using a ($2 \times 2 \times 1$) W (001) surface model, $1\,\mathrm{ML}$ of O atoms is absorbed on the top site of the surface, followed by doping rare earth atoms (La, Ce, Y) into the vacancy sites of the W-O lattice. The work functions of the system with rare earth atom coverages of $0.5\,\mathrm{ML}$ and $1\,\mathrm{ML}$ were calculated. Through liquid phase synthesis, plasma discharge sintering, and heat treatment, nano-scale second phase rare earth oxides(La_2O_3, CeO_2, Y_2O_3, etc.)-tungsten cathodes were produced. Different ignition experiments were designed to simulate various operating conditions. The cascade arc plasma source was used for mass-loss and lifetime prediction tests on the cathode materials. After testing, Scanning Electron Microscopy and Energy Dispersive Spectrum microscopic characterizations of the cathode materials were conducted to analyze their composition, morphology, and elemental distribution. Optimization results reveal that the W-La, W-Ce, and W-Y cathodes prepared with this method exhibit excellent ablation resistance and plasma bombardment endurance at high temperatures. The nanoscale dispersion of the doped phases endows the cathode with superior electron emission properties, enhancing the overall efficiency of the system. Under plasma density of $1.0 \times 10^{19}\,\mathrm{m}^{-3}$ and working temperature of $2000\,\mathrm{K}$, the projected lifetime of rare earth tungsten alloy cathodes exceeds $2000\,$hours.

Keywords: Field emission cathode, Rare earth tungsten alloy, First-principles, calculations, Work function.

I. INTRODUCTION

As the primary and neutralizing electron source of various electronic vacuum devices and spacecraft potential control systems, the performance of the cathodes not only affects the overall efficiency of the system, but is also the most important factor limiting long service life and high reliability of the system [1, 2]. Compared with various conventional thermal cathodes, field emission cathodes have a series of advantages, such as fast startup, room temperature operation, no preheating delay, and high current density, which has been applied in many fields such as electron beam lithography, vacuum diodes and space propulsion systems, and other fields [3]. At present, in the field of vacuum devices, the commonly used cathode materials are Ba-W cathodes, lanthanum hexaboride(LaB_6) and C12 A7 cathode materials, as well as new cathode materials developed on the basis of this, and the advantages of the application of different emitter materials are different [4–7].For example, in the Magneto Plasma Dynamic Thruster(MPDT), the cathode, as a core component, is in the center of the high-energy plasma plume, with an instantaneous ignition voltage of up to several thousand volts, an operating temperature higher than 1500K, and an extremely harsh working environment, which puts forward high requirements for the cathode material and structure [8].

In order to improve the cathode working life and efficiency, based on the traditional cathode electron emission theory and traditional cathode material types, this paper will carry out research from the perspective of cathode emitter material optimization.

Atomic models were constructed with tungsten(W)-O surfaces doped with various rare earth atoms (La, Ce, Y), using first-principles calculations and density functional theory(DFT). Calculations for the work functions were conducted for the models of $0.5\,\mathrm{ML}$ and $1.0\,\mathrm{ML}$ doping levels. The results showed that doping rare earth elements greatly lowered the work function of the alloy cathode, improving electron emission performance, and that $0.5\,\mathrm{ML}$ doping in W-O lattice sites resulted in the lowest work function.

In the present study, nano-doped rare earth tungsten cathode materials were prepared using liquid-phase synthesis and plasma discharge sintering techniques. A series of ignition tests on the thruster prototypes were conducted along with microstructural characterization experiments. Electron emission performance, ignition performance, and efficiency of a tungsten alloy cathode doped with various elements and proportions were tested.

In addition, based on the working principle of cathode for various vacuum devices, in view of the existing experimental conditions, the long-life test of the whole machine is subject to greater constraints. Therefore, this study independently conducts a life assessment experiment for cathode, and conducts a mass-loss-life prediction of cathode through a self-developed cascade arc plasma generator source. The results show that the nano rare-earth tungsten alloy cathode has better electron emission performance than the conventional cathode, in which the lanthanum oxide doped tungsten alloy with different mass fractions makes the cathode material escape work reduced significantly; after the independent life test of

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the cathode, the preliminary prediction of the rare-earth tungsten alloy cathode life reaches 2000 h.

II. ELECTRON EMISSION PRINCIPLE OF FIELD EMISSION CATHODE

The emission of cathodes used in thruster systems primarily relies on the principles of thermionic emission and field emission [9]. Thermionic emission follows the Richardson equation:

$$j_0 = [AT_K^2]exp(-\phi_k/kT_K) \tag{1}$$

As shown in Equation(1), j_0 represents the zero-field emission current density. A is the theoretical value of the material's emission constant, T_K is the cathode operating temperature, and ϕ_k represents the material's work function [10].

Similar to the theoretical derivation of the thermionic emission equation, Fowler and Nordheim developed the field emission theory for metals. They assumed the following: (1) the distribution of band electrons conforms to the Fermi-Dirac distribution; (2) a smooth, planar metal surface is considered, ignoring atomic-scale irregularities; (3) classical image forces affecting electrons are taken into account; (4) the work function distribution is uniform. Under these assumptions, the following equation holds:

$$j_0 = \frac{1.54 \times 10^{-6} \xi^2}{\phi_k} \exp\left[-\frac{6.83 \times 10^7 {\phi_k}^{3/2}}{\xi} \theta\left(y_0\right)\right] \tag{2}$$

In Equation(2), ξ represents the electric field strength, measured in V/cm.Where $\theta(y_0)$ is a slow-variable function of ξ,

$$y_0 = \left(3.79 \times 10^{-4} \frac{\sqrt{\xi}}{\phi_k}\right) \tag{3}$$

According to the electron emission equations above, the zero-field emission current density j_0 is closely related to parameters such as the material's work function, temperature, and electric field strength. Theoretically, the lower the work function of the cathode material, the more readily electrons within the material can overcome the surface potential barrier to emit from the cathode surface. Additionally, the operating temperature T_K and electric field strength ξ are directly proportional to the zero-field emission current density j_0 of the cathode. At lower temperatures, thermionically emitted electrons that overcome the barrier contribute negligibly to the emission current, with the emission primarily consisting of field-emitted electrons near the Fermi level [11].

For the work function of cathode materials, the periodic arrangement of lattice ions is interrupted at the metal-vacuum boundary, thereby disrupting the periodicity of the potential field [12]. The potential energy increases in a specific manner and approaches zero at infinity, forming the surface potential barrier of the metal. When electrons move to the metal surface and attempt to escape, they are hindered by this surface barrier, which is defined as the material's work function.

$$\phi_{\mathrm{k}} = E_{\mathrm{V}} - E_{\mathrm{F}} \tag{4}$$

All electrons attempting to escape from the metal must have energy at least equal to the Fermi level plus the value of the work function and follow a statistical distribution. Their average energy equals 32 KT, with each degree of freedom contributing an average energy of 12 KT, consistent with the results of kinetic molecular theory [13].

On the other hand, the cathode evaporation rate increases sharply with rising cathode operating temperature. Cathode evaporation directly impacts the cathode's lifespan, grid emission, and inter-electrode insulation performance. Ideally, a cathode should have high emission capability, requiring a low work function and minimal evaporation. Considering these two requirements, a quality factor F can be used to represent the performance:

$$F = \phi_{\mathrm{k}} \times 10^3/T_{\mathrm{e}}(eV/K) \tag{5}$$

Te is the temperature (K) at which the material's vapor pressure reaches 10^{-5} mmHg. To ensure thruster performance, the cathode temperature should not exceed its "vapor pressure temperature" Te [14].

Therefore, the selection and optimization of cathode emitter materials need to balance electronic emission performance with thermodynamic properties. Higher electron emission performance can enhance cathode discharge efficiency and overall thruster efficiency, while better thermodynamic properties extend the service life of the cathode under extreme operating conditions.

III. FIRST-PRINCIPLES STUDY ON THE SURFACE WORK FUNCTION OF RARE EARTH TUNGSTEN ALLOY CATHODE

Quantum mechanics is an important foundation of modern physics and one of the greatest discoveries of the 20 th century. Using quantum mechanics, it is possible to explain and predict the physicochemical properties of a wide range of systems and to quantitatively analyze the laws of their electronic motion. The first principle is a computational method based on quantum mechanics to study the properties of materials from the point of view of electron motion. The wave function contains all the information of the computational system, which greatly limits the scope of its practical applications, and the establishment of the density functional theory solves the problem of the complexity of the wave function. The basic idea of density functional theory is to change the characteristics based on the orbital wave function, with the particle density function to express the system base state of each physical quantity, to the electron density function represents the system energy [15, 16].

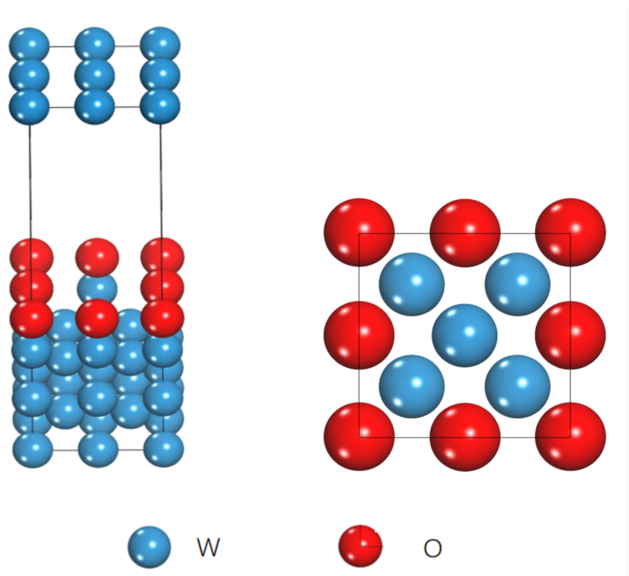

Fig. 1. Top-Site Adsorption of O Atoms on W(001) Surface.

Material Studio material simulation software incorporates a variety of three-dimensional scale simulation calculation methods, which can complete the cross-scale scientific research from the microscopic electronic structure to the macroscopic performance prediction [17]. It is on the basis of this advantage that the atomic-scale emission structure was modeled with the use of the Materials Studio in the current work. Geometric optimization was made for tungsten alloy models doped with various rare earth elements. Further on, the relaxation of surface atoms was conducted and work function was calculated under convergence conditions. The pseudopotential method was realized for solution of Schrödinger equation, while computations of work function were performed in LDA functional and PBE-GGA functional. The work function of the (001) crystal plane for tungsten is calculated in order to assess the possible effect the doped elements have on the current density of the cathode emission [18].

The models mainly include the following: the adsorption of O atoms and La atoms on a tungsten surface, adsorption of O atoms and Ce atoms on a tungsten surface, and adsorption of O atoms and Y atoms on a tungsten surface.

The current work has used the CASTEP density functional calculation module of Materials Studio, which is based on a plane-wave basis set. Among the well-known classical algorithms in CASTEP, the main one is the plane-wave pseudopotential method. By moving the model of a tungsten atomic structure and geometric optimization, it was possible to find the ground state with the lowest energy. We used the default number of maximum steps, while the cutoff energy was $278.0\,\mathrm{eV}$. The method of calculation was "Fine" and for the rest of the parameters that were given, we kept them the same as the default. Under the ultrasoft pseudopotential, GGA was being applied, and the functional from Perdew-Burke-Ernzerhof was picked to describe the electron exchange-correlated interactions [19].

O atomic layer was adsorbed on the surface of the supercell $W(2\times2\times1)$. According to the related computational literature, the larger the adsorption energy is, the more stable the adsorption system is. In fact, the adsorption energies of O atoms on the top site, bridge site, and hollow site of W supercell are about $9.11\,\mathrm{eV}$, $7.40\,\mathrm{eV}$, and $8.20\,\mathrm{eV}$, respectively; this means the O atoms are preferentially adsorbed on the top site shown in Figure 1 [20].

On the W-O (top-site) surface, rare earth atoms with varying coverages were adsorbed. Similar to O atoms, rare earth atoms on the $(2\times2\times1)$ W(001)-O (top-site) surface also have three possible adsorption positions. Due to the larger atomic radius of rare earth elements, adsorption at the top and bridge sites causes significant lattice distortion in the W lattice. Computational results indicate that rare earth atoms are more likely to adsorb at hollow sites on the W-O (top-site) surface. The formula for calculating the adsorption energy of rare earth atoms is as follows:

$$E_{ad} = -\frac{1}{N}(E_{La+W(001)-O(top)} - NE_{La} - E_{W(001)-O(top)}) \tag{6}$$

As shown in Figure 2a to 2c, the work function for rare earth atoms in the W-O crystal with a coverage of $0.5\,\mathrm{ML}$ was calculated using Equation (6). Because rare earth atoms readily lose their two outermost valence electrons, transferring them to the inner O atoms, the electron density of the coverage layer is lower than that of the substrate surface layer. This results in a dipole layer with a positive charge on the outside, raising the surface potential and reducing the barrier height.

Similarly, this study further calculated the work function of W-O crystals with a rare earth atom coverage of $1.0\,\mathrm{ML}$, meaning that rare earth atoms fully occupy the hollow sites in the W crystal, as shown in Figure 2d to 2f.

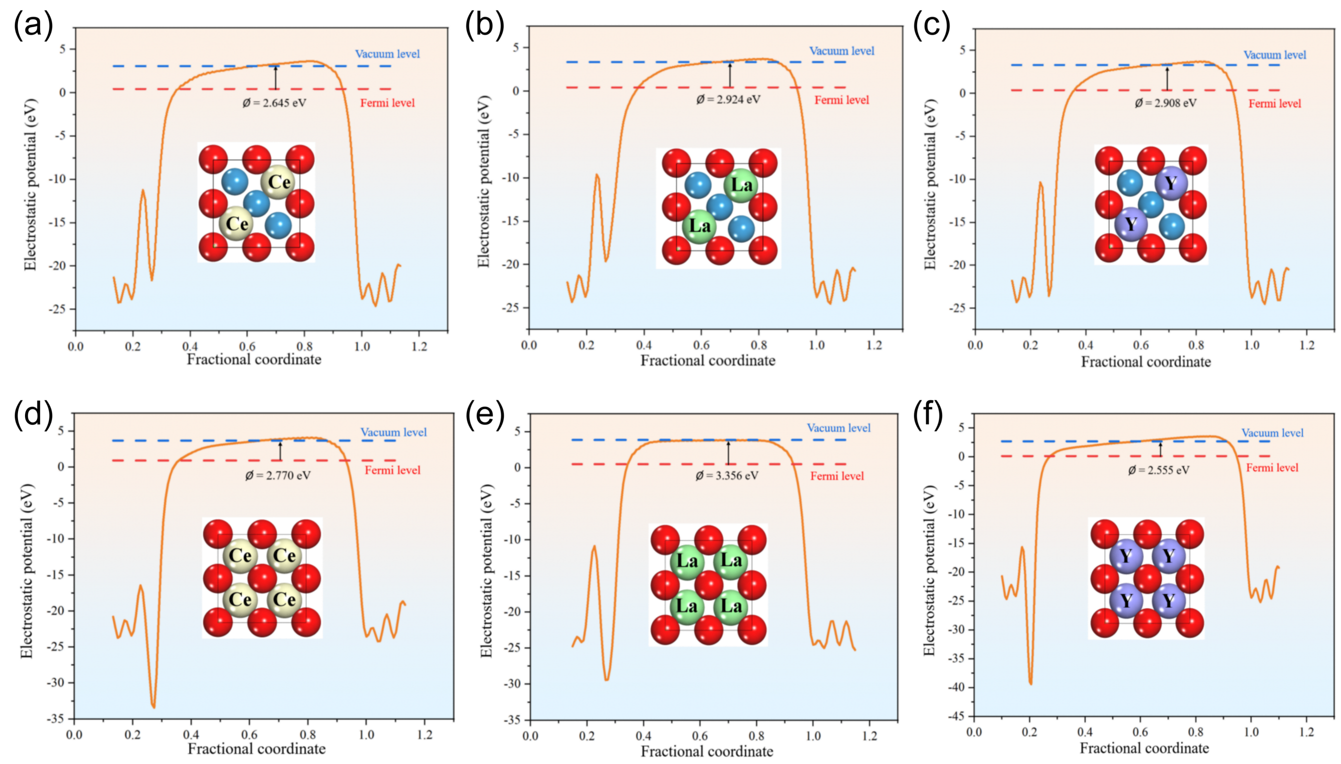

Fig. 2. W-O Crystal Surface Doped with rare earth atoms model and work function calculation.(a) Doped with 0.5 ML Ce; (b) Doped with 0.5 ML La; (c) Doped with 0.5 ML ML Y; (d) Doped with 1.0 ML Ce; (e) Doped with 1.0 ML La; (f) Doped with 1.0 ML Y.

Table 1. Work Function of W-O (Top-Site) Doped with Different Rare Earth Atoms.

W(001)-O Top-Site Doping	Work Function (eV)
0.5 ML Ce atoms	2.645
0.5 ML La atoms	2.924
0.5 ML Y atoms	2.908
1.0 ML Ce atoms	2.770
1.0 ML La atoms	3.356
1.0 ML Y atoms	2.555

As marked in Table 1, the calculational results of work function for W-O (top-site) doped with 0.5 ML and 1.0 ML of different rare earth atoms show that the large atomic radius of rare earth elements results in large lattice distortion when fully occupying the hollow sites in the W crystal, and therefore such a system is unstable. When the number of adsorbed atoms increases above each optimal coverage, the interactions between dipoles increase gradually. Here, the middle atoms are depolarized by an electric field of adjacent dipoles. The reduction of a dipole moment increases the work function.

Comparing the data in the table, doping rare earth atoms into the W crystal reduces the surface work function in all cases, validating the feasibility of this study's approach to optimize cathode electron emission performance by doping tungsten with rare earth elements, thereby enhancing the efficiency of magnetoplasma thrusters. Additionally, the calculation results show that doping 0.5 ML La or Ce atoms at the W-O (top-site) achieves the greatest reduction in work function. Due to the relatively smaller radius of Y atoms, full Y atom doping into the hollow sites of the tungsten crystal results in minimal lattice distortion, and its work function is slightly reduced compared to 0.5 ML Y doping.

IV. EXPERIMENTAL STUDY ON CATHODE OF RARE EARTH TUNGSTEN ALLOY

A. Synthesis and processing of rare earth tungsten alloy cathodes

For various vacuum electronics and space propulsion systems, the primary requirements for cathodes are superior electron emission capability and high ablation resistance to withstand impacts from high-energy particles. Experimental research indicates that the electron emission performance, melting point, and ablation resistance of rare earth tungsten alloy cathodes are closely related to the chemical properties, physical dispersion, and percentage content of the nano-doped phase. We synthesized W-La_2O_3 alloy cathodes with different doping levels and studied their electron emission performance at various temperatures and voltages for different W-La_2O_3 doping concentrations [21].

Figure 3a shows the preparation process of the nano rare earth tungsten alloy cathode. Using liquid-phase synthesis and reduction, nano rare earth tungsten alloy cathode ma-

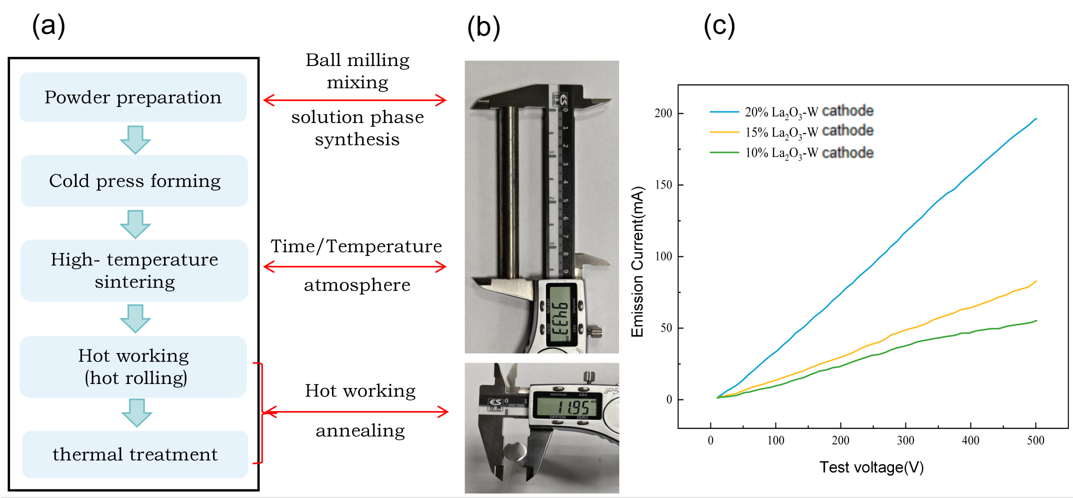

Fig. 3. Processing workflow of rare earth tungsten alloy cathodes and electron performance testing of W-La cathodes.

terial precursor powder was prepared, with rare earth oxide particles well-dispersed in the tungsten matrix. The higher the dispersion uniformity of the second-phase rare earth oxides, the more balanced the ablation process at the cathode tip, resulting in lower ablation levels [22–24]. The mixed powder is then processed into standard rod cathodes through cold pressing, high-temperature sintering, hot working (hot rolling), and heat treatment, as shown in Figure 3b, which displays a W-La cathode.

The series of rare earth tungsten alloy cathodes we designed and processed are aimed at the extreme working environment of vacuum equipments and space thrusters. By optimizing the emitter composition, we aim to balance high electron emission performance with ablation resistance for extended service life. Figure 3c shows electron emission performance data for W cathodes doped with various mass fractions of W-La_2O_3. As W-La_2O_3 content increases, the overall electron emission performance of the cathode improves significantly, confirming that adding low work function components to optimize emitter performance is feasible.

In addition, we prepared W-Ce and W-Y rare earth tungsten alloy cathodes with different doping ratios using the same processing method and made preliminary predictions of their properties [25, 26].

B. Independent service life experiment using cascade arc plasma source

Under conditions of existing experimental possibilities, the verification of the service life of cathodes is a long-time and expensive process, and even more expensive are tests of life validation conducted together with thrusters. As space missions have imposed longer lives on electric propulsion systems, full-life ground testing has become increasingly impractical. For instance, the ground-tested ion thruster for NASA's *Deep Space 1* mission lasted for 30 352 hours, which is more than five years. In comparison, the JIMO mission would have used six ion thrusters powered by nuclear energy as its main propulsion; these would have to individually be rated for 83,000-hour lives. Assuming an efficient test duration of 75 percent, a 1.5-times redundancy life test would take approximately 19 years [27].

We have developed a cascade arc plasma source to simulate the real working environment of rare earth tungsten alloy cathodes. This consists of a vacuum system, the power supply system, the superconducting magnet system, the plasma generation device, the gas supply system, the water cooling system, and the Langmuir probe system, as shown in Figure 4a.

In this paper, it is considered that the tip morphology of the cathode changes significantly when the mass loss of the cathode reaches 10% - 15%.This reduces the effectiveness of small-hole current limitation and leads to inability to maintain stable discharge when the propellant flow rate exceeds the set operating range, and the cumulative operating time under rated conditions cannot be achieved. Beyond this point, the cathode is to be considered functionally degraded and at the end of its service life.

Therefore, as shown in Figure 4c to 4e, we used our custom cascade arc plasma source to simulate the thruster's experimental environment by placing the cathode within the plasma source and setting specific plasma density and temperature parameters. Material ablation and service life predictions were conducted based on mass loss over a specified experimental duration. This study performed independent lifetime experiments and comparisons for W-La cathodes, W-Ce cathodes, and pure W cathodes [28].

The mass loss of the three experimental cathode materials under the conditions of 3 h experimental duration with the plasma density up to $1.0 \times 10^{19}\,\mathrm{m}^{-3}$ measured by Langmuir probe and the temperature up to 1300°C measured by the bottom plate temperature probe is shown in Figure 4b. Based on parameters such as gas flow rate, input current, magnetic field strength, and plasma density, the service life of the W-La cathode is estimated at approximately 3000 hours, and for

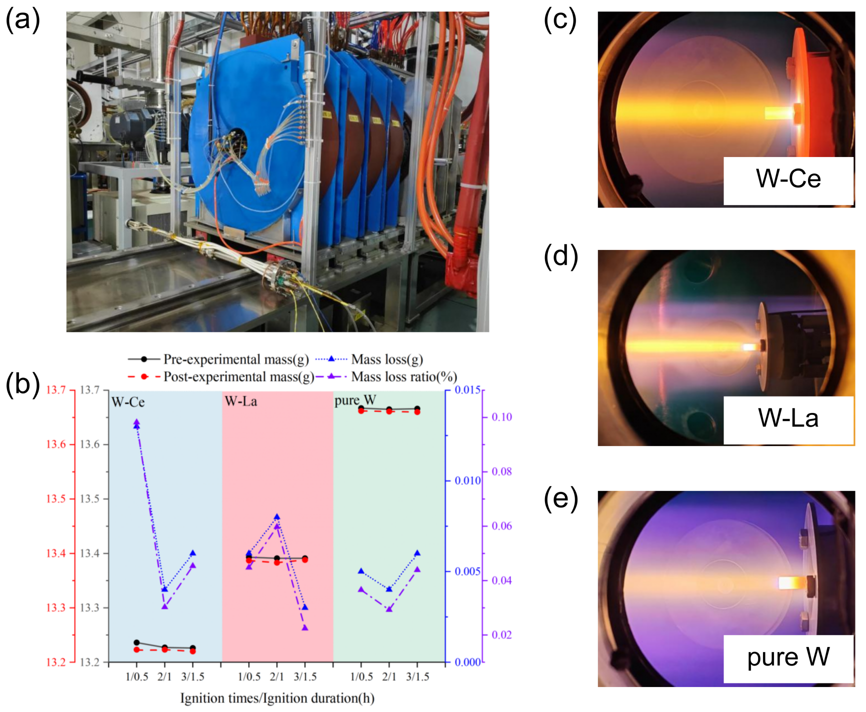

Fig. 4. Ablation life prediction experiment by cascade arc plasma generation source system.(a) Cascade arc plasma generation source system(Institute of Plasma Physics, Chinese Academy of Sciences, China); (b) Test cathodes mass loss data chart; (c) W-Ce cathode test; (d) W-La cathode test; (e) Pure W cathode test.

the W-Ce cathode, about 1100 hours. This reduction is attributed to decreased thermal resistance of the cathode matrix as rare earth element content increases, leading to higher mass loss under extreme operating conditions and consequently reduced service life.

Thus, in optimizing the performance of cathodes, it is essential to ensure both excellent electron emission performance and high ablation resistance.

C. Microstructural characterization of rare earth tungsten alloy cathodes

The melting, sputtering, and eventual deposition on the surface of the cathode material due to the W matrix are critical factors affecting the lifespan and efficiency of the cathode material. Preliminary analysis of the deposits shows no new elements, consisting solely of tungsten oxides, which exhibit valence changes at high temperatures [29].

The nominal operation tests of the ignition demonstrated severe ablation at the tip for the rare earth-doped tungsten alloy cathodes. SEM images show that the mm-sized holes at the tip of the cathode have contracted several times due to high-temperature ablation during operation, as shown in Figure 5.Leading to a very irregular emitter surface that significantly influences propellant flow there at the cathode tip. This results in more difficult ignition breakdown with the increase of ignition duration and frequency under the same power conditions.

The W-La cathode after the experiment was further analyzed by EDS energy spectrum, as shown in Figure 6. Among them, Figure 6a is the W-La tip deposit, and the analysis results show that the content of W is as high as 65.7%, followed by O element, and the content of rare earth La is as low as 1.6%, indicating that La is doped into the W matrix in the form of W-La_2O_3, which is consumed during the discharge process to form electron emission. The analysis of particulate matter on the cathode surface in Figure 6b shows that it is the oxide of W, and the matrix of W melts and recrystallizes at high temperature, changing valence and forming different oxides of W.

The characterization results show that the rare earth oxide W-La_2O_3, due to its low work function, allows electrons near the Fermi level of La to overcome the surface barrier and emit from the material under high voltage between the cathode and anode. It results in the appearance of pores, which can be distributed on the surface of emitters in a very uneven manner, and it also demonstrates that the poor work function given by rare earth elements has been wildly added in order to improve the total performance and efficiency of emitters. Moreover, the density, size, and evenness of such pores on the surface depend on many factors, including the size of the second phase particle, the doping mass fraction, and the raw material mixing methods.

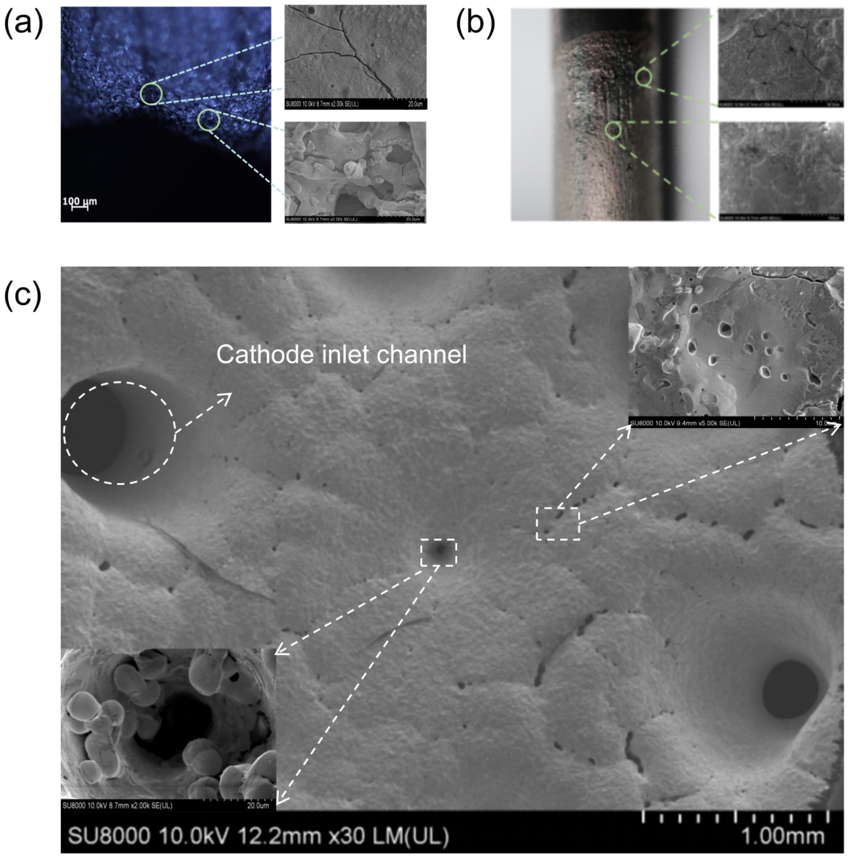

Fig. 5. Scanning Electron Microscopy (SEM) of W-La Cathode after the Experiment.(a) Tip area; (b) Transition area and normal area; (c) Tip hole area.

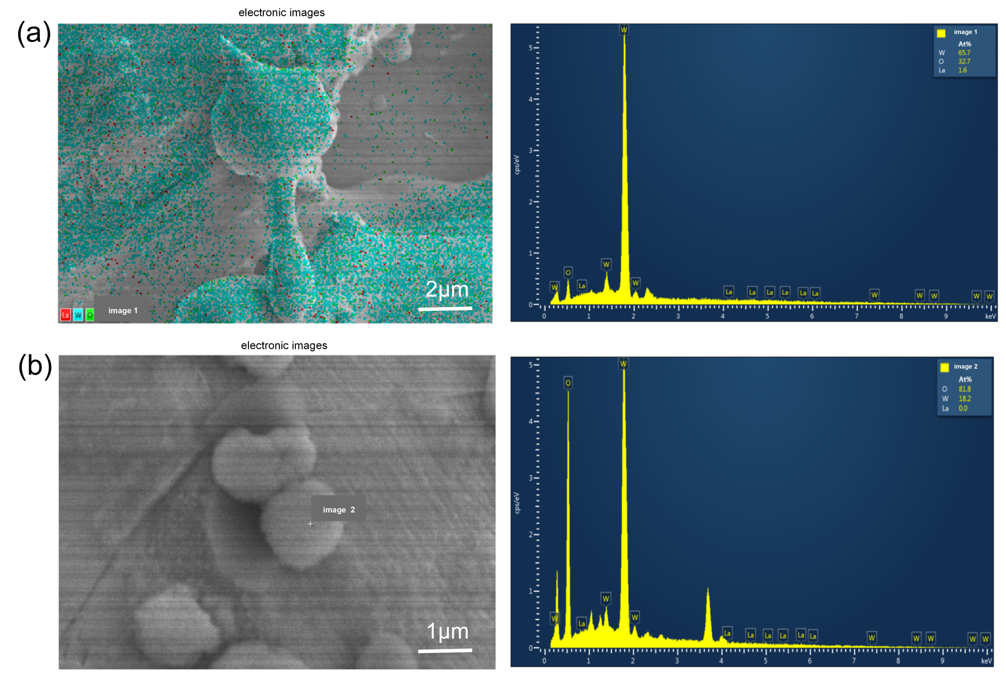

Fig. 6. Energy Dispersive Spectrum (EDS) of W-La Cathode after the Experiment.(a)Area scanning; (b)Spot scanning.

V. RESULTS AND DISCUSSION

The cathode is one of the crucial components of various electronic vacuum devices and space propulsion systems, and the electron emission performance and ablation resistance

of cathode materials against high-energy particle impacts directly affect the overall efficiency, performance and life of the system and other key indicators [30]. Therefore, the future research on cathode needs to consider the doping elements affecting the electron escape work of cathode materials, thus affecting the electron emission performance of cathode, in addition, the ratio of doping elements and W matrix affects the melting point of the overall alloy, that is, it affects the sputtering resistance of cathode, etc.; in addition, the preparation method of cathode also greatly affects its performance, and the optimization of the cathode preparation process enables the dopant phase to form a nano-sized in W matrix. dispersion, the better the dispersion, the better the improvement effect on the cathode emission performance [31].

In this paper, the optimization of high performance field emission rare earth tungsten alloy cathode is investigated by both simulation and experimental verification.Atomic models are built with Material Studio for rare earth elements adsorbed onto the W-O (top-site) surface; the relevant surface work functions are then calculated based on density functional theory. The work function values calculated were $2.645\,\mathrm{eV}$, $2.924\,\mathrm{eV}$, and $2.908\,\mathrm{eV}$ for $0.5\,\mathrm{ML}$0.5 ML of Ce, La, and Y adsorbed on the W-O top site and $2.770\,\mathrm{eV}$, $3.356\,\mathrm{eV}$, and $2.555\,\mathrm{eV}$for $1.0\,\mathrm{ML}$ of Ce, La, and Y, correspondingly. These results reflect that doping of rare earth atoms effectively reduces the surface potential barrier of the cathode, which confirms that doping of rare earth atoms is an effective method to enhance performances of electron emission for cathodes.

Of course, based on the Material Studio simulation result, in the paper, W-La, W-Ce, and W-Y cathodes with different treatment methods were prepared; a serial of ignition tests have been designed and performed; the results indicate that compared to pure tungsten cathodes, the operating condition for the rare earth tungsten alloy cathode operating is more stable with lower ignition voltage. A cascade arc plasma source custom's development was also employed to simulate the real working environment that tungsten alloy cathodes face. On the basis of all the same quantity of these operational parameters, namely, the gas flow rate, input current, magnetic field strength, and plasma density, the service lives for the considered rare earth tungsten alloy cathodes were estimated.

Optical microscopy, SEM, and EDS were conducted on the cathodes postexperiment. The experimental results obtained show that the role of the rare earth doped in the tungsten alloy cathodes is to excite the outer valence electrons at high voltage and break down the propellant to form plasma. In continuous electron emission, the consumption of the rare earth atoms is gradual. However, with the increase of the doping ratio of rare earth, to a certain extent high-temperature resistance of the cathode declines and serious ablation occurs. Thus, by changing the ratio of doping atoms in the cathode, an optimization of the trade-off between excellent electron emission performance and good resistance against ablation can be achieved.

Based on the above research on different rare earth tungsten alloy cathodes, we believe that high-performance field emission rare earth tungsten alloy cathodes will have great application prospects in many aspects such as vacuum devices, space propulsion, etc., in which the excellent electron emission performance of tungsten-lanthanum cathode can greatly improve the emission efficiency of the cathode itself. The ablation mechanism of rare earth tungsten alloy cathode and the development of longer-life cathode will be the next research focus.

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